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# Soft X-ray Studies of Pu Electronic Structure: Past Lessons and Future Directions

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## Soft X-ray Studies of Pu Electronic Structure: Past Lessons and Future Directions

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Photoelectron Spectroscopy (PES) [1] and X-ray Absorption Spectroscopy (XAS, Figure 1) [2-4] have contributed greatly to our improved understanding of Pu electronic structure. From these and related measurements, the following has been determined.

1. The Pu 5f spin-orbit splitting is large.
2. The number of Pu5f electrons is near 5.
3. The Pu 5f spin-orbit splitting effect dominates 5f itineracy.

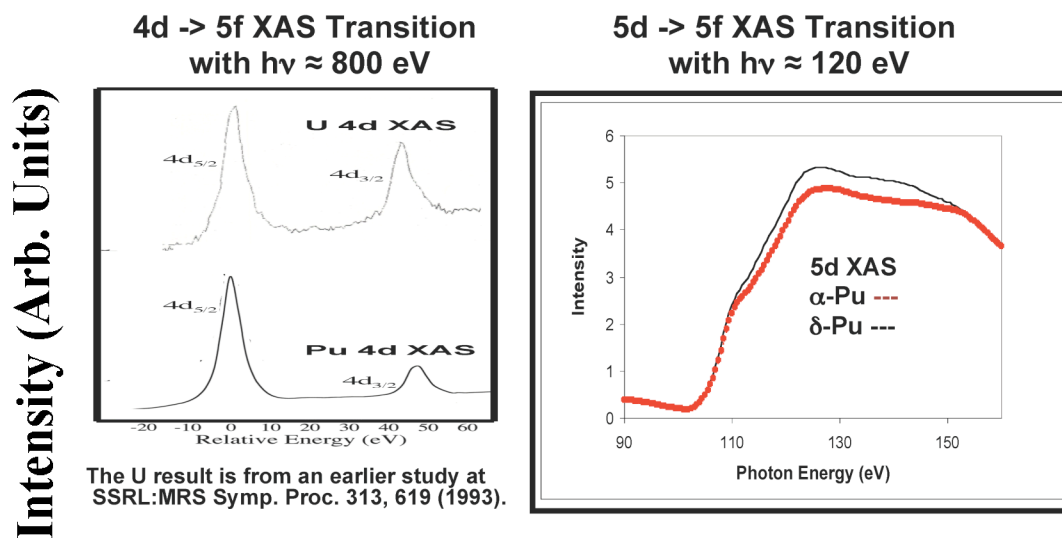


Figure 1

Significant questions remain concerning the nature of Pu electronic structure. Perhaps the missing piece of the puzzle is the direct experimental determination of the unoccupied electronic structure using high energy inverse photoelectron spectroscopy or Bremsstrahlung Isochromat Spectroscopy (BIS). [5] Past BIS studies of Th and U indicate the feasibility and utility of Pu studies. [6]

To this end, a new BIS capability has been developed in our laboratory, as shown schematically in Figure 2 below. [7]

## LLNL Fano and BIS Spectrometer

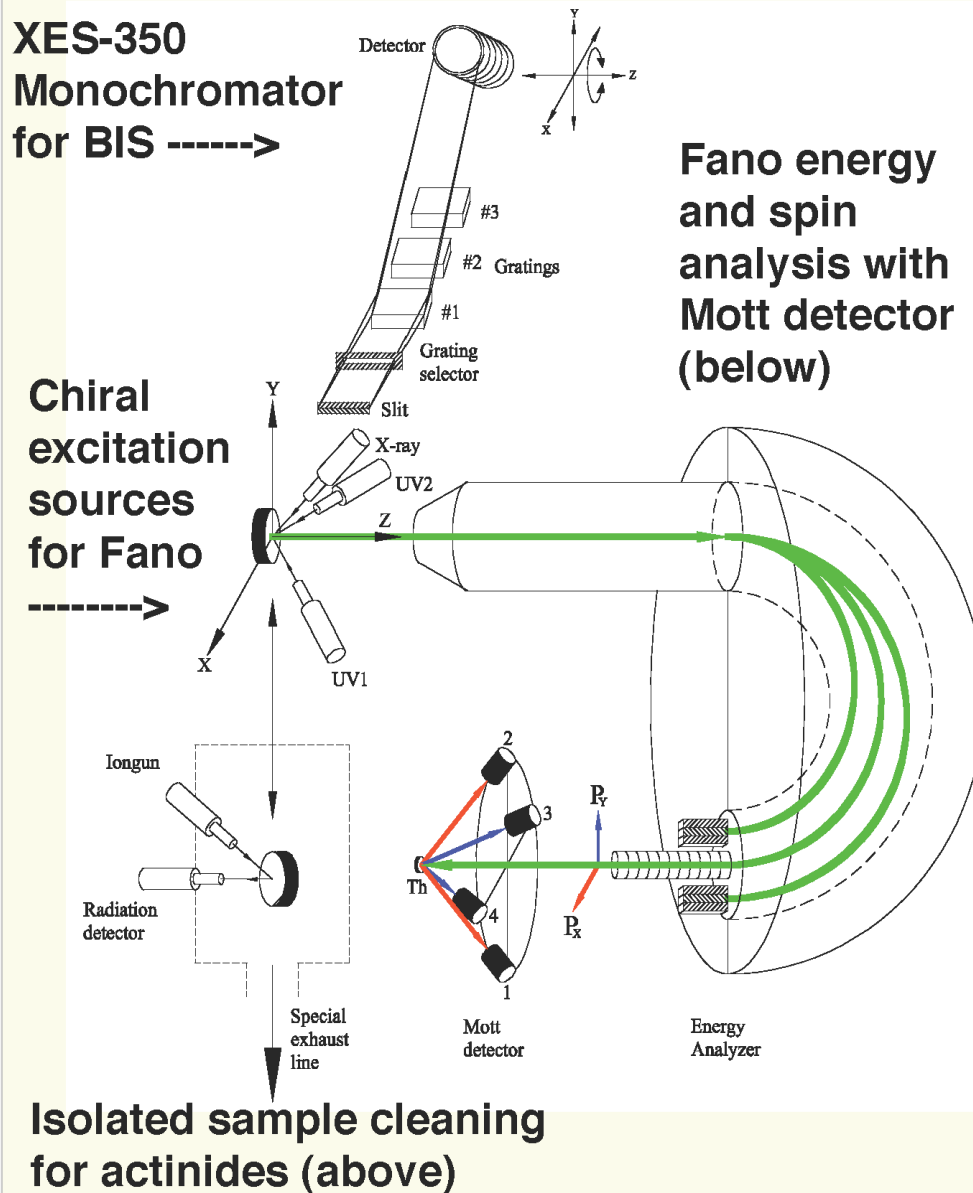


Figure 2

Electron stimulated emission of photons has been carried out using the XES-350 monochromator and detector system. Some of our preliminary results are shown below, using an electron excitation beam energy of 3000 eV.

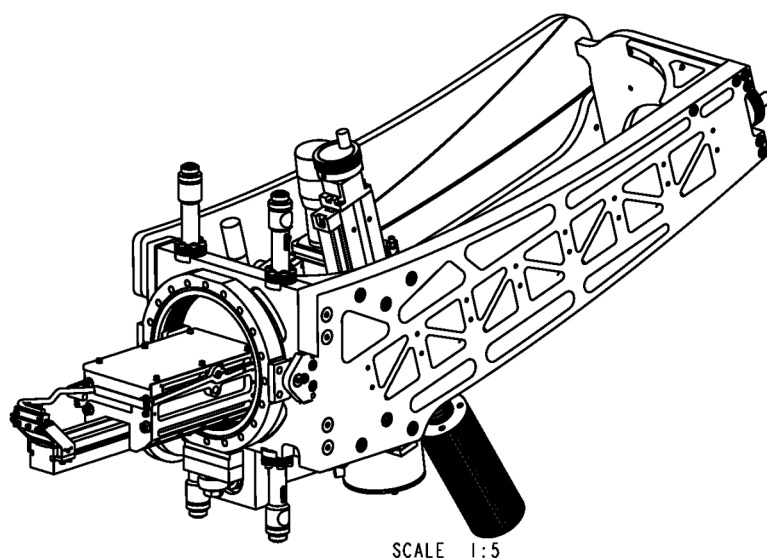
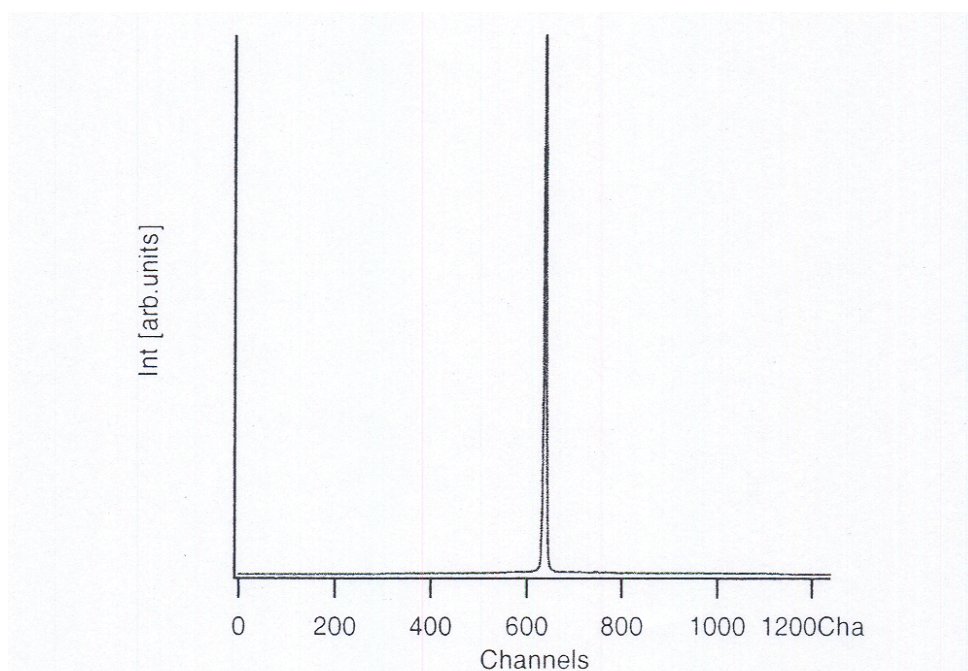


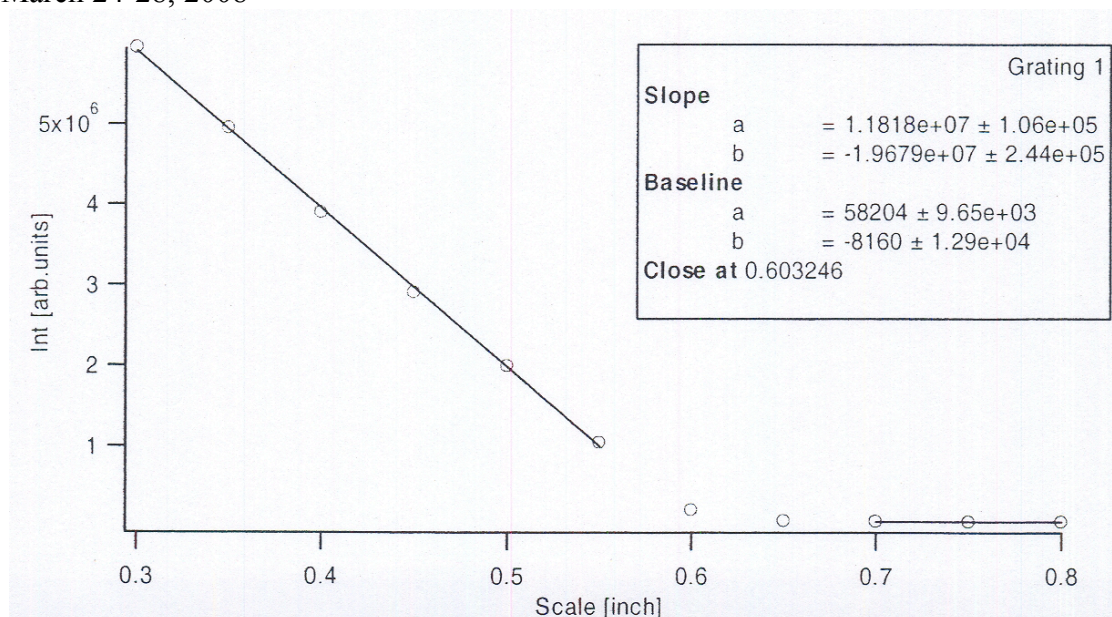
Figure 3  
XES-350 A Schematic of the monochromator and detector is shown here. Figure provided by Scienta.



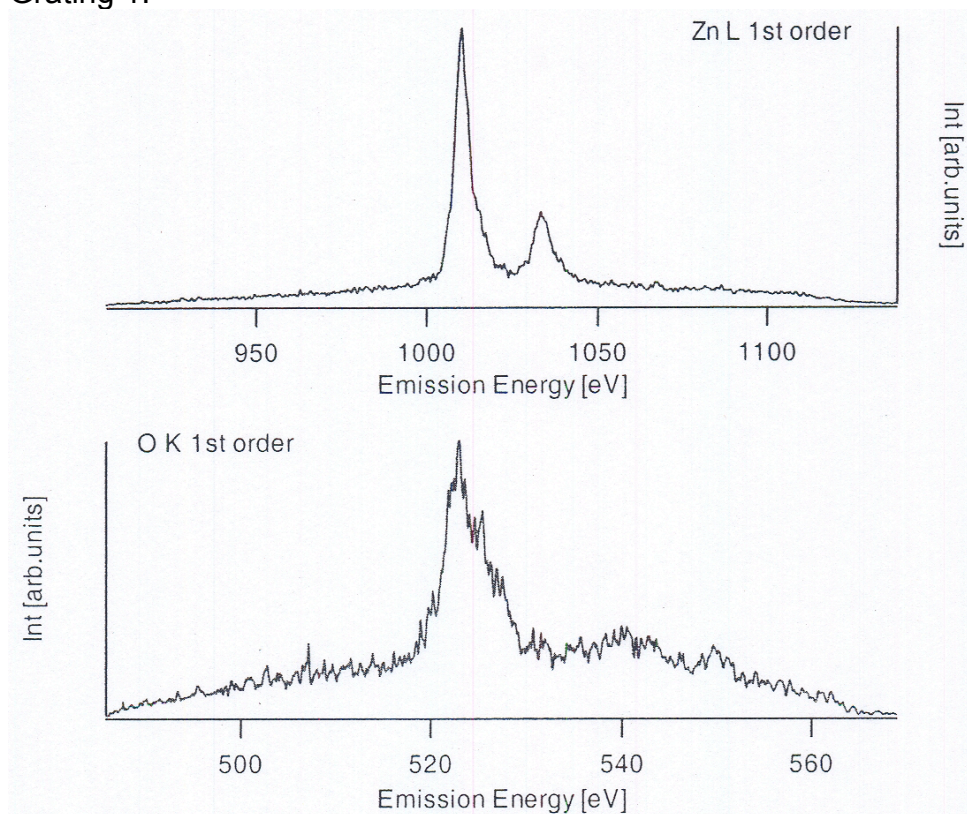
The 0<sup>th</sup> order peak with a 10  $\mu\text{m}$  slit at the focus position. The FWHM is  $\sim 5$  channels.

Figure 4





**Figure 5**  
 The intensity of the transmitted radiation as a function of the slit inch scale for Grating 1.



**Figure 6**  
 Zn L (2p) and O K (1s) emission in first order, measured at the calculated positions on Grating 1.



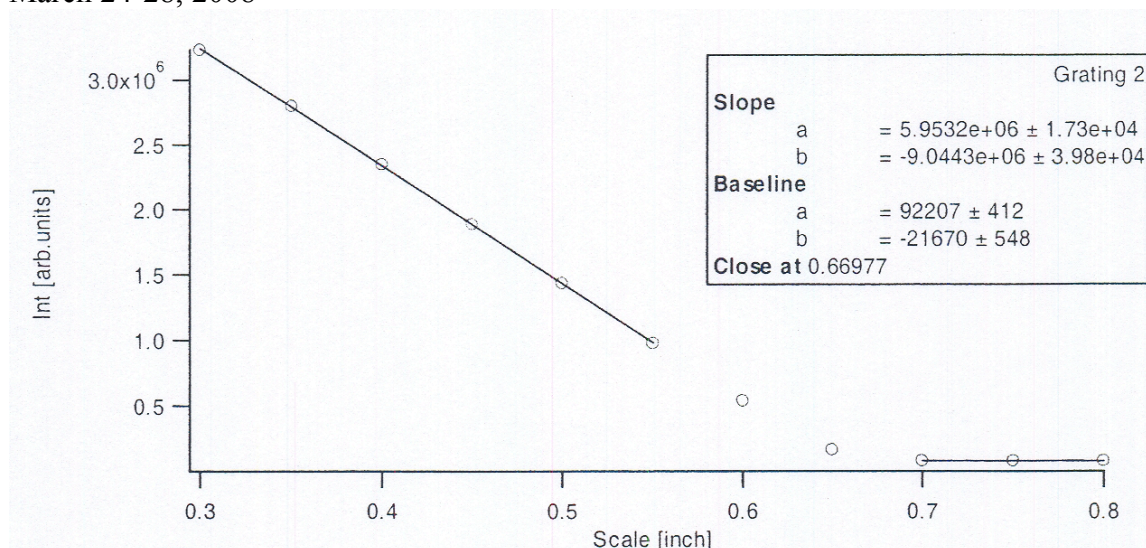


Figure 7  
 The intensity of the transmitted radiation as a function of the slit inch scale for Grating 2.

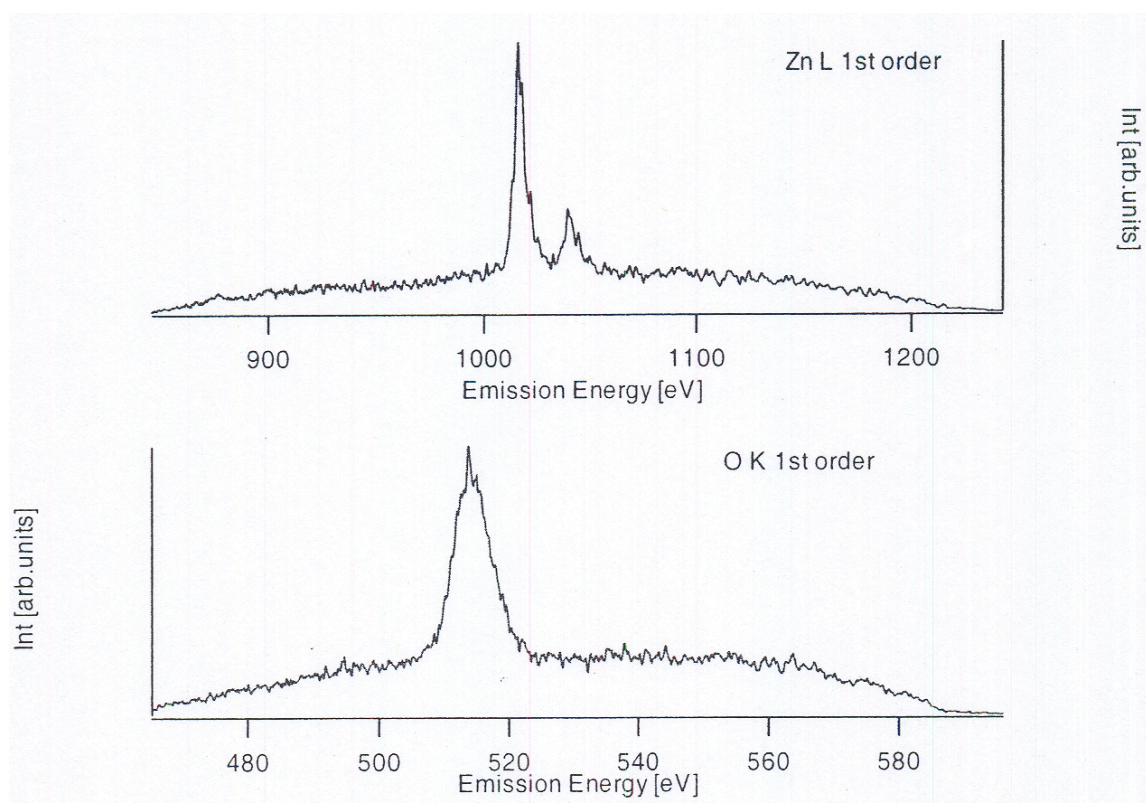
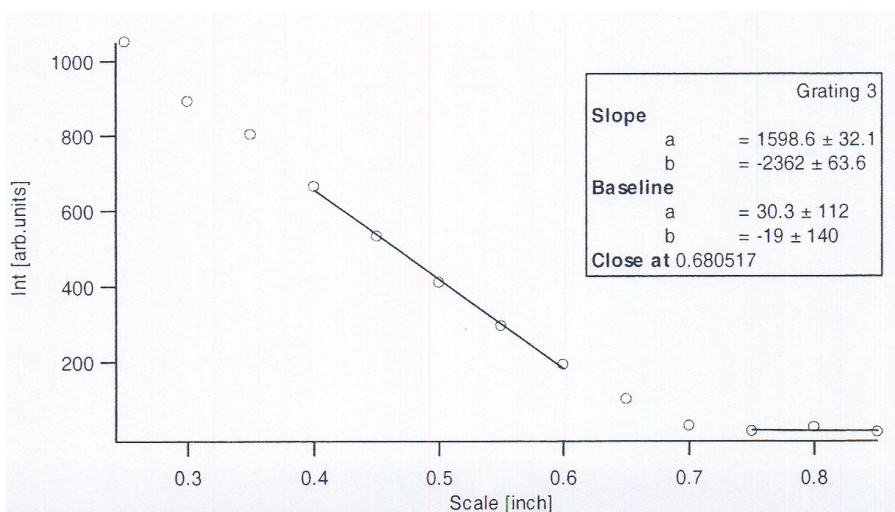


Figure 8  
 Zn L (2p) and O K (1s) emission in first order, measured at the calculated positions on Grating 2.



**Figure 9**  
 The intensity of the transmitted radiation as a function of the slit inch scale for Grating 3.

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#### References

1. J.G. Tobin et al, Phys. Rev. B **68**, 155109 (2003).
2. K.T. Moore et al, Phys. Rev. Lett. **90**, 196404 (2003).
3. G. van der Laan et al, Phys. Rev. Lett. **93**, 097401 (2004).
4. J.G. Tobin et al, Phys. Rev. B **72**, 085109 (2005).
5. J.G. Tobin, M.T. Butterfield, N.E. Teslich Jr., R.A. Bliss, M.A. Wall, A.K. McMahan, B.W. Chung and A.J. Schwartz, "Using Nano-focussed Bremsstrahlung Isochromat Spectroscopy (nBIS) to Determine the Unoccupied Electronic Structure of Pu," in "Recent Advances in Actinide Science," Royal Society of Chemistry, ed. R. Alvarez, N.D. Bryan and I. May, page 773 (2006).
6. Y. Baer and J.K. Lang, Phys. Rev B **21**, 2060 (1980).
7. J.G. Tobin, S.W. Yu, T. Komesu, B.W. Chung, S.A. Morton and G.D. Waddill. MRS Symp. Proc. **986**, 63 (2007), Document # 0986-OO01-08.